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published in

Modern Methods and Algorithms of Quantum Chemistry,
J. Grotendorst (Ed.), John von Neumann Institute for Computing, Jülich, NIC Series, Vol. **2**, ISBN 3-00-005746-3, p. 44, 2000.

http://www.fz-juelich.de/nic-series/

Dissipative quantum dynamics for laser induced desorption

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A new approach[1] to dissipative quantum dynamics shall be applied in the theoretical description of laser induced desorption of small molecules from metal oxide surfaces. Wave packet calculations on ab initio potential energy surfaces for the electronic ground state and a representative excited state have been performed for NO adsorbed on a NiO(100) surface[2, 3]. However, the finite lifetime of the excited state has been included only semi-empirically.

To treat the dissipative quantum dynamics rigorously, we employ the surrogate Hamiltonian theory[1]. The adsorbate/substrate complex is divided into the system and the environment. The bath consists of electron-hole-pairs in the surface which are described as two-level-systems. The adsorbate is coupled to the bath via a dipole-dipole-interaction and transitions between the ground and excited potential energy surfaces are induced by this coupling. A surrogate hamiltonian with a finite number of bath modes is constructed leading to a controllable approximation in which the necessary number of bath modes depends on the propagation time. The laser pulse exciting the adsorbate is included semiclassically. Thus the influence of different pulse lengths, intensities and shapes on the desorption as well as the interplay of dissipation and excitation can be investigated.

We present the method and first results.

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